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Direct Measurement of Aircraft Engine Soot Emissions Using a Cavity-Attenuated Phase Shift (CAPS)-Based Extinction Monitor

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The optical properties of soot particles in plumes emanating from a high bypass turbofan aircraft engine (V2527) were measured at distances of 40–80 m behind the engine with a cavity-enhanced phase shift (CAPS)-based extinction monitor (known as the CAPS PM_{ex}) and a multi-angle absorption photometer, both operating at wavelength ∼630 nm. Integrated plume measurements from the two instruments were highly correlated with each other ($r^2 > 0.99$, N = 12) and with measured carbon dioxide emission concentrations. Ancillary measurements indicated that the soot particle volume-weighted mobility diameter distribution peaked at 60 nm with a full width at half maximum of ∼60 nm. The soot single scattering albedo determined using the absorption and extinction measurements under engine idle conditions was 0.05 ± 0.02 (where the uncertainty represents 2σ precision), in agreement with previous measurements of aircraft exhaust. The engine soot emission index (mass soot per mass fuel burned) for this particular engine, derived from these measurements and a wavelength-specific mass absorption coefficient and the measured in-plume carbon dioxide concentrations, was 225 ± 35 mg kg^{-1} at engine idle conditions. These results plus more limited data collected from in-use aircraft on the runway indicate that the CAPS extinction monitor can provide (with an appropriate albedo correction) a credible measurement of the engine soot emission index in situations where the time response and sensitivity of particle absorption monitors are not otherwise sufficient.

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INTRODUCTION

The impact of aircraft gas turbine engine emissions upon the atmosphere has drawn extensive attention in recent years because of the predicted increase in air traffic (Fahey et al. 1995; Schlager et al. 1997; Anderson et al. 1998; Paladino et al. 1998; Schuman et al. 2002; Unal et al. 2005; Wey et al. 2007). While most of this attention has focused on the emission of greenhouse gases (specifically carbon dioxide), comparatively little effort has been spent on understanding the effect of aircraft-generated soot (i.e., black or light absorbing carbon) on climate change (Petzold et al. 1999). Light absorbing carbon has both direct and indirect effects on the atmosphere; it strongly absorbs sunlight, thereby warming the ambient air, which leads to further net warming, but can also promote cloud formation (and thus potential net cooling) by providing sites for water droplet. The effect of emitted soot at ground level is an area of intense activity because high concentrations of soot have been linked to adverse health effects (ACRP 2008).

Soot emission levels are typically derived from measuring the absorption of light by soot particles and converting that value to mass by dividing by an assumed mass absorption coefficient (Bond and Bergstrom 2006). There are two types of commercially available particle absorption monitors: the filter-based monitors (e.g., particle soot absorption photometer, aetholometer), and those that directly measure absorption using photoacoustic techniques (e.g., photo-acoustic soot spectrometer). Of the commercially available filter-based instruments that have been used for the detection of soot emitted from aircraft engines (Hagen et al. 1996; Brock et al. 2000; Agrawal et al. 2008; Onasch et al. 2009), the multi-angle absorption photometer (MAAP) (Petzold et al. 2002) has been perhaps the most successful in obtaining data during aircraft emission tests. It relies on the collection of soot particles on a glass-fiber filter substrate and measures the resultant change in light attenuation; uniquely for filter-based instruments, it also makes a real-time correction for scattered light. The manufacturer's precision for the determination of light absorbing carbon (or soot) is <0.1 μg m^{-3} with an integration time of 120 s. However, like its brethren, it

requires a substantial sample flow rate to achieve high sensitivity or short sampling times, is prone to matrix effects, and utilizes a derivative measurement to provide instantaneous concentration values. While these limitations are inconsequential when soot measurements are undertaken either using an engine located on a test-stand or in close proximity to engines affixed to a stationary aircraft, there are situations where the availability of an instrument that provides greater sensitivity at a far lower sample flow would be advantageous. These include monitoring aircraft plumes while in-flight and in this particular circumstance, monitoring engine exhaust plumes of operating aircraft as they take off, land, and taxi on the runway.

The photoacoustic approach, which measures aerosol absorption in free flow, was developed in order to overcome a number of these deficiencies. The research instrument recently developed by researchers at National Oceanic and Atmospheric Administration (NOAA), when operated at 532 nm, demonstrates a much better sensitivity than any filter-based instrument, <1 Mm^{-1} at 1-s integration time (corresponding to <0.2 μg m^{-3} of soot) when operated at a maximum response time of 5 s (D.A. Lack 2011, personal communication). However, its noise level rises rapidly if the flow rate is increased sufficiently to provide 1-s time response, a problem that is endemic to this type of detection technique. The only commercially available photoacoustic device, the Photoacoustic Soot Sensor (PASS), when operated at 781 nm, demonstrates a level of detection (LOD, 3σ) of $\sim$1 Mm^{-1} (<0.3 μg m^{-3} of soot) with an integration time of 10 s; if operated at 532 nm, its LOD rises to $\sim$13 Mm^{-1} (Flowers et al. 2011). We also note that the accuracy of photoacoustic particle spectrometers at high relative humidity has been questioned (Murphy 2009). To our knowledge, there is only one study applying the photoacoustic technique to the measurement of aircraft engine exhaust (Rogers et al. 2005).

In order to achieve greater sensitivity and faster time response than filter-based instruments under these circumstances, we investigate the use of a newly available particle extinction monitor (CAPS PM_{ex}) as a means of measuring soot concentrations. The proposed use of particle extinction monitor as a surrogate for a proper absorption measurement makes a critical assumption: particle extinction and absorption are same within reasonable approximation – i.e., size-averaged soot particle single scattering albedo (SSA) approaches zero. In order for this to be true, the soot particles (with any organic coatings, if present) must be small. Typical particle mass distributions of soot generated from modern airplane gas turbine engines peak at mobility-based diameters well below 100 nm under all operating conditions (i.e., both idle and takeoff), a finding which has been shown for the CF6, CF34, CFM, PW2037, and JT8D engines (Herndon et al. 2008), the F404 engine (Rogers et al. 2005), and the CFM56, AE3007A, PW4158, and RB211 engines (Kinsey et al. 2010) over a wide range of operating conditions, a sampling that is representative of the major aircraft engine manufacturers (Pratt and Whitney, Rolls Royce, and General Electric). Given a measured absorption coefficient of soot and the calculated (Mie) scattering cross sections, the expected average soot SSA for these engines is well below 0.1. In fact, the only reported measurement of the SSA of aircraft engine-generated soot (Petzold et al. 1999) indicates that the value is below 0.05. Moreover, these small particles should not exhibit any nonlinear optical effects; i.e., particle absorption is proportional to particle mass. Recent measurements by Cross et al. (2010) demonstrate that bare soot particles in this diameter range indeed show linear absorption behavior with respect to mass.

As part of an ongoing collaboration under the Airport Cooperative Research Program (ACRP) to study gaseous and particulate matter (PM) emissions from various modern aircraft (ACRP 2008), we present results of measurements of airplane engine-generated soot obtained at distances of 40–80 m behind the engines using a particle extinction monitor based on cavity attenuated phase shift (CAPS) measurement techniques (Kebabian and Freedman 2007; Kebabian et al. 2007). We also present a limited amount of soot emission data from operating aircraft while the instruments were stationed alongside an operating runway. The CAPS technique, similar in nature to cavity ringdown, relies on the use of a sample cell employing high reflectivity mirrors. In this particular application, square-wave modulated red light ($\sim$630 nm) from a light emitting diode (LED) is directed through one mirror and into the sample cell. The distortion in the square wave caused by the effective optical path length within the cavity ($\sim$1 km) is measured as a phase shift in the signal as detected by a vacuum photodiode located behind the second mirror. The presence of particles in the cell causes a change in the phase shift (ϑ), which is related to the total extinction (the sum of scattering and absorption), $\varepsilon_{\text{part}}$ by the following relationship:

$$\cot\vartheta - \cot\vartheta_0 = \frac{c}{2\pi f}\varepsilon_{\text{part}}, \qquad [1]$$

where ϑ_0 is the phase shift measured in the absence of particles, c is the speed of light, and f is the modulation frequency. The CAPS PM_{ex} extinction monitor, when using red light, has a detection level of less than 2 Mm^{-1} (<0.3 μg m^{-3}) with a time response of 1 s using a flow of only 0.85 L/min and thus is highly suitable for the detection of short duration (5–60 s) emission plumes. Complete details of this extinction monitor and its performance are presented elsewhere (Massoli et al. 2010).

EXPERIMENTAL

On February 18, 2010, dedicated engine tests involving an airplane (with two V2527 engines) provided by United Airlines were performed at Chicago O'Hare International Airport. Emission plumes of the aircraft were investigated using a variety of gaseous and particle measurement instruments located on board of the Aerodyne mobile laboratory (Herndon et al. 2005). The particle measurement instruments employed during

these tests pertinent to this study included an MAAP (Thermo Environmental), which employs a LED centered at ~630 nm, the CAPS-based particle extinction monitor (PM_{ex}) (Aerodyne Research), which employs a LED centered at 630 nm, a condensation particle counter (CPC, Model 3776, TSI), and an engine exhaust particle sizer (EEPS, Model 3090, TSI). These instruments provided information about particle absorption and extinction, number density, and mobility-based size distribution. A high-resolution aerosol mass spectrometer (HR-AMS, Aerodyne Research) provided information about the possible presence of any semi-volatile coatings on the soot. Carbon dioxide concentration measurements were provided by a Li-Cor 820 carbon dioxide analyzer and total NO_x (the sum of NO and NO_2) concentration was measured using a chemiluminescence-based analyzer (Model 42I, Thermo Environmental) operated in single channel mode with 2-s time resolution.

During this entire test period, the airplane was parked in front of the United Airlines maintenance facility; the engines were operated at known fuel flow rates indicative of near-idle conditions. The Aerodyne mobile laboratory, located ~40–80 m behind the aircraft, was continuously driven in and out of the engine exhaust plumes during the 50-min study. Given the relatively low sensitivity of the MAAP, it sampled the plume at a flow rate of ~15 Lpm through its own 12.5 mm o.d. sampling line; all the other instruments shared another sampling line with a total flow of ~16 Lpm, the preponderance of which was used by the EEPS device. A cyclone (Model URG-200–30EH, URG) with a 2.5 micron diameter cutoff was employed to remove large particles on both sampling lines.

The CAPS PM_{ex} monitor supplies its own filtered purge air, which prevents particles from fouling the mirrors. Both pressure and temperature of the gas within the cell are measured and used to subtract the contribution to the total extinction from gas phase Rayleigh scattering. One complication is that the PM_{ex} monitor is also sensitive to the presence of absorbing gases in the plume, in this case nitrogen dioxide. Since there was no direct measurement of the NO_2 concentration in the sampled plumes, total NO_x was used as a surrogate species. A correction factor was determined by measuring the response of the extinction monitor to NO_2 within the plume by flowing the air sample through a filter that removed all particles and then proportioning that number to a separate measurement of total NO_x provided by the chemiluminescence-based analyzer. This NO_2 correction factor was then scaled directly with the NO_x measured in the plume while monitoring soot particles. This correction thus assumes that the ratio of NO_2 to NO_x is constant when the engine is operating in the idle mode. In any case, this correction is quite small, decreasing the measured SSA by 0.01, a value well within the precision, much less the accuracy, of the subsequent measurements.

On the same day during another period of time, the Mobile Laboratory was parked on a vehicular service road located parallel to both an active taxi-way (at a distance of ~100 m) and runway used for take-offs (at greater distance). The Mobile Laboratory was parked such that it and its sampling probe pointed into the wind (with velocity of ~5–7 m s^{-1}) at all times. A video camera was used to record all airplanes that passed the Mobile Laboratory as well as any vehicles that passed the Mobile Laboratory on the service road. The delays between visual sighting and emission plume detection varied between 15 and 60 s depending on whether the plume originated from the taxi-way or the takeoff runway.

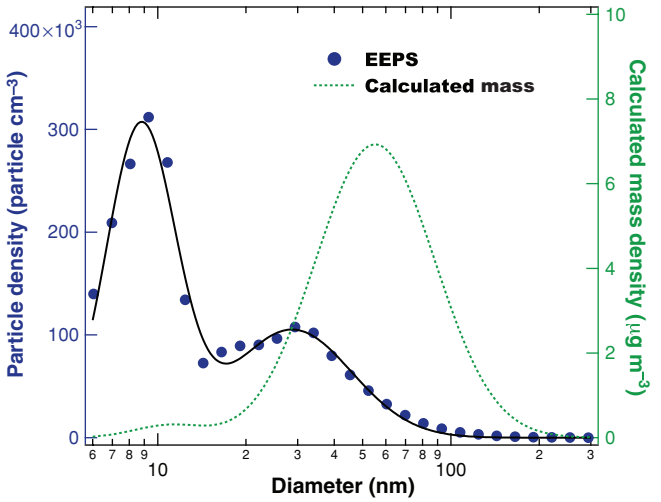

FIG. 1. Plot of typical measured particle concentration (left;, labeled EEPS) and calculated mass density (right) versus particle mobility diameter. (Color figure available online.)

RESULTS AND DISCUSSION

Characterization of the V2527 Engine

Exhaust Particle Size Distribution

In this study, particle size number density distributions of PM from the exhaust plumes were determined using the EEPS during each plume measurement and fitted with a bimodal log-normal function in order to resolve nucleation and soot modes (Onasch et al. 2009). A typical spectrum is shown in Figure 1; all the collected size distributions during these studies were virtually superimposable, indicating that, at least under engine idle conditions, the soot emission size distribution was not affected by the relatively short mixing times (a few seconds) between emission and detection. The nucleation particles, which peak in size around 8 nm, are dominant in the total particle count. It is believed that these nucleation particles consist of semi-volatile species such as sulfates (Curtius et al. 1998; Pueschel et al. 1998; Karcher et al. 2000) and organics (Onasch et al. 2009; Timko et al. 2010), which contribute little to the scattering component of the extinction signal (~1 Mm^{-1} assuming that they are spherical and the Mie theory applies). Furthermore, sulfate particles do not absorb at 630 nm and in any case represent a very small fraction of the total mass; on the other hand, a second volume-weighted component that peaks at ~60 nm (mobility diameter) and becomes negligible at nominal diameters

greater than 100 nm particles is attributed to combustion soot. Soot particles are known to be fractal-like aggregates (Bonczyk and Hall 1991; Koeylue et al. 1995; Cross et al. 2010) with a mass-mobility exponent of ~2.0, but a simple Mie scattering calculations under the assumption of sphericity provides an estimate of the expected contributions of absorption and scattering. Using the complex refractive index of soot suggested by Bond and Bergstrom (2006) (m = 1.95–0.79 i), one obtains a theoretical average albedo of ~0.03 for the expected size distribution. This indicates that the extinction measurement should be dominated by the absorption of light by the soot.

One potential complication is that depending on engine power and other conditions, a certain amount of the emitted semi-volatile organic and sulfate material in the engine exhaust will eventually coat the emitted soot (Herndon et al. 2005; 2008; Onasch et al. 2009). These potential coatings can have two effects on the optical properties of the soot particles. The first is on extinction, where the coating, either by increasing the size of the particle or changing its fractal dimension, could increase the scattering cross section and increase the total extinction. The second is that the apparent absorption is enhanced by a lensing effect caused by a layer of non-graphitic material (Bond et al. 2006; Lack et al. 2009; Shiraiwa et al. 2009; Cross et al. 2010). In this study, the Aerodyne HR-AMS was used to directly monitor the presence of semi-volatile coatings on soot particles (Jayne et al. 2000; Jimenez et al. 2003; Drewnick et al. 2005). We found that the mixing ratio of semi-volatile PM organic to black carbon was approximately 10% within a factor of two. Onasch, et al. (2009) had previously demonstrated that for aircraft engine-generated soot with a similar soot volume-weighted mass size distribution, a determination of non-carbonaceous mass fraction can be made using the aerosol mass spectrometer. Even if all this organic material (or even a factor of 2–3 times the measured value) were found as a coating on the soot, one would expect little effect on either extinction or absorption at this low level of contamination. Similar conclusions were reached by Fuller et al. (1999).

Soot Particle Single Scattering Albedo (SSA) Determination

A portion of the time series of MAAP and CAPS PM_{ex} particle measurements during the trial period are presented together in the bottom panel of Figure 2; the top panel presents the carbon dioxide concentration measurements over the same period. Integrated values for the particle absorption, particle extinction, total NO_x, and carbon dioxide were obtained for each of the measured plumes. Integrating the plumes instead of providing a point-by-point ratio removes any effects caused by differences in time response and provides a number of distinct measurements that can provide a reasonable measure of experimental precision. All plume measurements were corrected for the existence of ambient background levels.

The integrated values for optical absorption and extinction (after correction for the presence of NO_x) are presented as a correlation plot in Figure 3. The results indicate an extremely high correlation ($r^2 > 0.999$) between the MAAP-based absorption and CAPS PM_{ex}-based extinction measurements over signal levels, which vary by a factor of 60. Furthermore, the

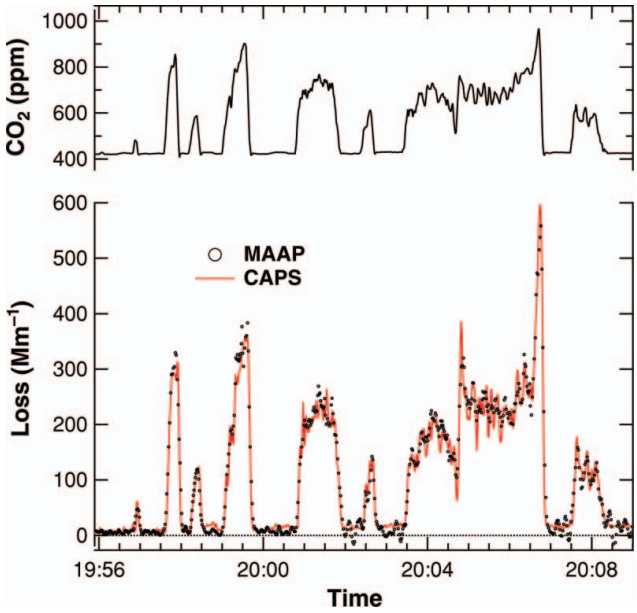

FIG. 2. Plots of particle optical extinction (labeled CAPS) and absorption (MAAP) (bottom panel) and carbon dioxide concentrations (top panel) measured in the air sample as the ARI Mobile Laboratory is driven in and out of the engine exhaust plume emanating from a fixed aircraft. (Color figure available online.)

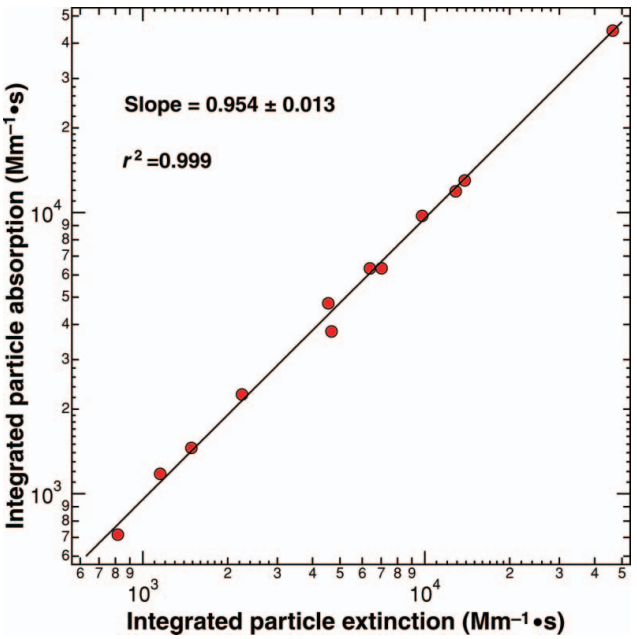

FIG. 3. Correlation plot of data taken using the MAAP (absorption) and CAPS PM_{ex} (extinction) monitors. The data are derived from the integrated plumes from the aircraft engine and are presented in units of loss × time. (Color figure available online.)

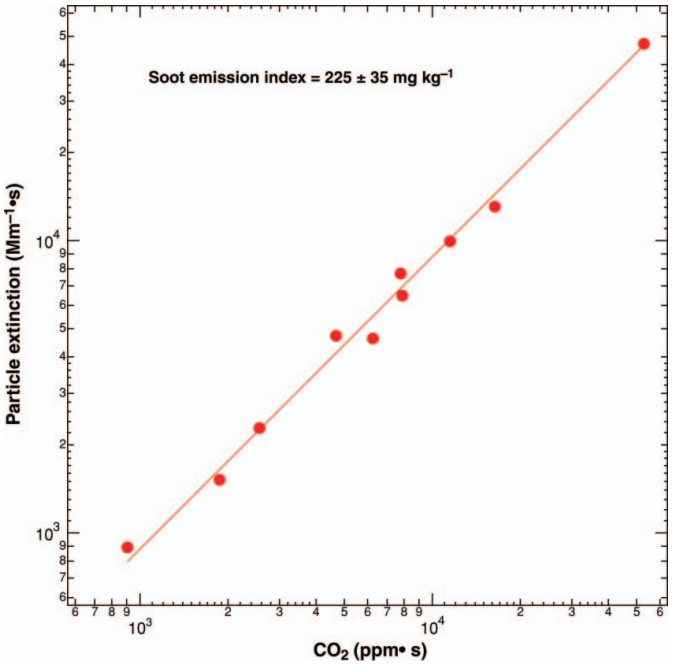

FIG. 4. Correlation plot of plume-integrated particle extinction versus carbon dioxide. Also shown is a linear least squares fit to the data. The soot emission index is calculated using the slope of the fitted line, the measured single scattering albedo and a mass specific absorption coefficient. (Color figure available online.)

obtained slope indicates that the soot SSA is approximately 0.05 ± 0.02, which agrees well with a previous measurement of aircraft engine exhaust of 0.035 ± 0.02 obtained using a particle soot absorption photometer (absorption) and nephelometer (scattering) (Petzold et al. 1999). It should be noted that this level of agreement is perhaps fortuitous given the level of accuracy (no better than $\pm 10\%$) that can be attributable to any of the instruments.

Engine Exhaust Soot Emission Index

A widely used measure of the contribution of engine exhaust soot to emission inventories is the emission index or mass soot emitted per mass fuel burned. As the measured soot extinction is a function of distance from the engine and local meteorological conditions (e.g., wind speed and direction), it must be proportioned to another species, such as carbon dioxide, which provides a metric of fuel consumption. We present in Figure 4 a correlation plot of the integrated plume extinction (from the CAPS PM_{ex}) versus the integrated carbon dioxide concentration. The correlation between these two quantities is excellent ($r^2 = 0.996$); using a linear fit to the data, one obtains a value of 0.88 ± 0.02 Mm^{-1} (extinction) ppm^{-1} (carbon dioxide concentration). In order to derive an emission index, the soot mass is calculated from the soot extinction by dividing the measured extinction (after correction for the measured SSA) by the mass normalized absorption cross section, 6.4 ± 0.5 m^2 g^{-1}, obtained at a wavelength of 633 nm (Mulholland and Choi 1998). The mass of fuel burned is obtained by converting the mole fraction of carbon dioxide to absolute carbon mass using the ideal gas law and then assuming that the nominal chemical formula of the fuel is $CH_{1.9}$ (Timko et al. 2010). Using this set of conversions, one obtains an engine soot emission index under engine idle conditions of 225 ± 35 mg kg^{-1} fuel burned. The quoted uncertainty is dominated by the estimated accuracy of the extinction measurement and the mass absorption coefficient. To our knowledge, this is the first such measurement of this engine.

Analysis of Emission Plumes from In-Use Aircraft

While runway emissions were sampled for a period of several hours, we present here a limited set of data meant to illustrate the potential of the CAPS PM_{ex} monitor as a means of measuring soot emission indices of aircraft under normal operating conditions. A more comprehensive analysis focusing on characterizing aircraft emissions will be presented at a later date. Figure 5 presents exhaust emission data for two airplanes

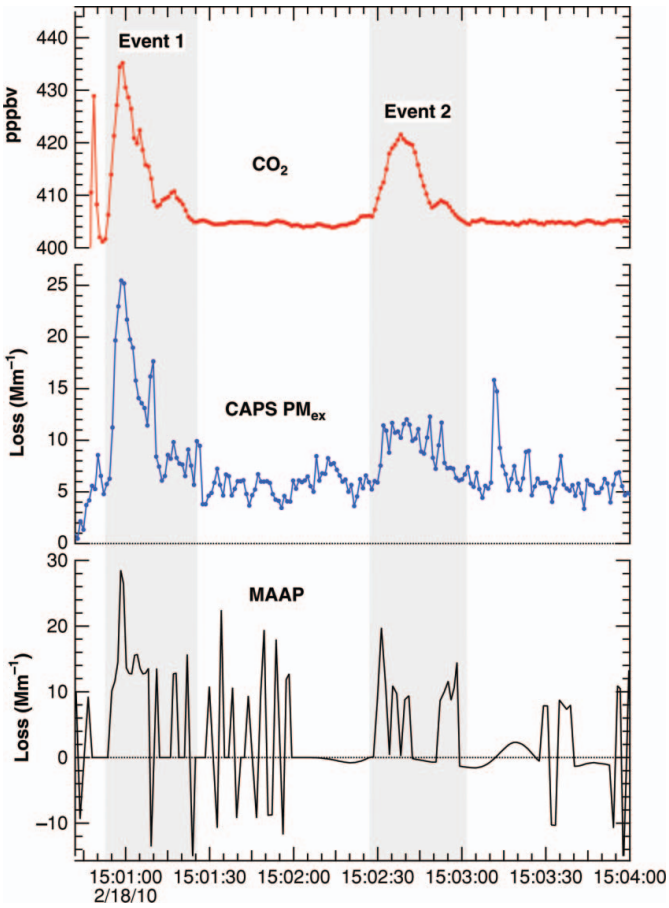

FIG. 5. Plots of carbon dioxide concentration (top), particle optical extinction (CAPS PM_{ex}; middle), and particle absorption (MAAP; bottom) measured in the air samples taken at a location alongside an operating runway. The plumes labeled Events 1 and 2 derive from engine emissions from airplanes in the process of accelerating for takeoff. (Color figure available online.)

(labeled as Events 1 and 2), both of which were in the process of taking off and thus presumably operating at near full throttle. During this period, no other airplanes appeared on the taxiway or on the runway. There was a constant delay of ~60 s between the time the airplane appeared in the center of the video camera frame and the arrival of the plume, indicating that at a wind speed of ~6 m s^{-1}, the plumes, which reached the intake port, derived from airplanes that were approximately 400 m distance from the Mobile Laboratory.

The emission plumes, Events 1 and 2 in Figure 5, were generated by a mid-size jet and a small commuter jet, respectively. Even for the larger magnitude plume, Event 1, the signals in all three instruments are much smaller in magnitude than that observed for the test data presented above and generally of much shorter duration. The difficulty of measuring particle absorption caused by the soot from engine emissions in this circumstance is amply demonstrated. Even so, the CAPS PM_{ex} is able to reproduce both the shape and time response of the carbon dioxide data for both Events 1 and 2 within the noise limitations of the instrument (± 1.5 Mm^{-1} [3σ]). The MAAP data, instead of being smoothed as in Figure 2, are presented as true 1-s data. As can be seen, the accuracy and precision of this data is limited by the digitization noise of the instrument, which is amplified by the need to take a derivative of the signal in order to provide the requisite time response. The only plume that is unequivocally observed by the MAAP is Event 1. Event 2 is observed if one integrates the data, but at a reduced level of probable accuracy.

For Event 1, the averaged CAPS PM_{ex} and MAAP data agree quite well with an observed albedo of 0.04 ± 0.2, where the large uncertainty is largely a function of the limitations of the MAAP. No attempt to calculate an observed albedo for Event 2 was made given the involvement of extremely large uncertainties. The soot emission indices for the engines involved in Events 1 (identified as a CFM56–7B24 engine) and 2 (CF34–8C5B1 engine) were determined to be (using the extinction monitor data and a SSA of 0.04) 130 ± 25 mg^{-1} kg^{-1} and 150 ± 35 mg^{-1} kg^{-1}, respectively, where the uncertainty represents an estimate of the precision with which the plumes can be integrated and compared. Both of these values are in line with the range of values determined by Herndon and co-workers (2008) in a campaign to measure runway-based takeoff emissions levels.

CONCLUSIONS

We have been able to demonstrate that a total optical extinction measurement of engine exhaust soot using the CAPS PM_{ex} particle extinction monitor provides complementary information to filter-based optical absorption monitors for the determination of emitted soot mass and thus soot emission index, especially in circumstances where available particle absorption monitors do not have the requisite time response and sensitivity. A comparison of the data obtained with a filter-based monitor (MAAP) with an extinction monitor (CAPS PM_{ex}) indicates that the size-averaged SSA of aircraft engine soot is quite small – ~0.05 – as expected from both calculations and previous measurements. The observation of negligible optical scattering in aircraft engine exhaust plumes, i.e., ~5% of total extinction, is a direct consequence of the soot particle volume-weighted size distributions, which peak below 100 nm and implies that particle extinction measurements can be used as surrogate absorption measurements. Furthermore, the CAPS PM_{ex} monitor provides high sensitivity and fast time response ($<.3$ μg m^{-3} [3σ] at 1-s sampling time) at much lower sample flow rates than any filter-based particle absorption monitor (e.g., MAAP, PSAP, or aetholometer).

The data from the fixed aircraft tests and runway observations indicate that employing the extinction monitor (especially in conjunction with current filter-based particle absorption monitors) when sampling aircraft engine emission plumes can expand the conditions under which reasonably accurate measurements are made. Measurements of the soot emission indices of in-use aircraft from idle to full throttle engine conditions in the process of taking off using the extinction monitor corrected for the measured SSA agreed well with previous data taken on fixed aircraft using a filter-based instrument.

This study shows the utility of CAPS-based particle extinction measurements in the context of characterizing the black carbon emissions from aircraft gas-turbine engines. As our current measurements are not exhaustive, we recommend deploying the extinction instrument in conjunction with a monitor that provides a direct absorption measurement.

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